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Physical pre-treatment of plums (*Prunus domestica*). Part 1. Modelling the kinetics of drying

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Abstract

An alternative physical method for enhancing the drying rate of plums is proposed. It consists of the superficial abrasion of the plums' peel using an inert abrasive material to remove the cuticular waxy layer, the limiting factor for moisture loss. The physical pre-treatment was compared with a chemical treatment in which the plums were dipped into a solution of ethyl oleate. The drying kinetics of the above samples, including the untreated one, were reconstructed by using a mathematical model. The drying process, carried out at 60 °C to reduce the prunes' quality loss, showed the great capability of both pre-treatments to enhance water diffusivity in the plum peel with respect to the untreated samples. Moreover, it was found that the physical treatment was more effective than the chemical one. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Abrasion; Dipping; Drying; Modelling; Prune

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1. Introduction

The prune occupies a small niche market, controlled almost completely by the USA and, more recently, by France; the development of production hinges on two factors: cost and product differentiation. The energy expended to dry plums constitutes about a quarter of the total cost of production (Bousignon, Cartier, Coquinot, Letang & Sardan, 1988; Sabarez & Price, 1999). Innovative action in the drying of plums centres on decrease in duration, and reduction of thermal damage, to produce a high quality product—a prune with good texture and bright colour. The desiccation plants generally use tunnels with carts in continuous movement counter to the airflow; the cycle is conducted for a period of 26–30 h at a temperature of 75–80 °C (Sansavini & Lugli, 1998). The epicuticular wax is the limiting factor for the plums' moisture loss; it consists of an underlying amorphous wax layer adjacent to the cuticle proper, together with crystalline granules of wax protruding from the surface (Price, Sabarez, Storey, & Back, 2000; Storey & Price, 1999). Different studies have been undertaken to reduce the times of desiccation, for instance direct osmosis as a pre-treatment to the desiccation in a current of warm air, and the pre-treatment performed with steam at 92–95 °C for 5 min, to remove the waxy layers of the fruit without lesions to the peel (Mastrocola, Pestalozza, dall'Aglio, & Lerici, 1990). Moreover, a diffusion model for prune dehydration was proposed by Sabarez and Price (1999).

In this paper, the effectiveness of a physical pre-treatment, consisting of the superficial abrasion of the plums' peel using an inert abrasive material to remove the cuticular waxy layer (the limiting factor for moisture loss), was evaluated. Therefore, plum dehydration (at 60 °C), of a diffused late cultivar, was studied by means of a mathematical model. The *Angeleno* cultivar, usually consumed fresh, was considered interesting for prune production, owing to its size and texture.

2. Materials and methods

Samples of *Angeleno*® plums (*Prunus domestica*) at commercial maturity were obtained from the public "Fruit Tree Research Institute" orchards in the Campania

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Nomenclature

c_1	Water concentration in the plum pulp (mol/m^3)
c_2	Water concentration in the plum peel (mol/m^3)
c_3	Water concentration in air (mol/m^3)
D_i	Water diffusivity in the grape pulp ($i = 1$) and peel ($i = 2$) $\left(\mathrm{m}^2/\mathrm{h}\right)$
h	Convective mass transfer coefficient (m/h)
H	Plum humidity, (%) dry matter
r	Distance from the plum centre (m)
R_0	Plum stone radius (m)
R_1	Pulp radius (m)
R_2	Overall plum radius (m)
t	time (h)
K_1	Equilibrium constant, defined by the ratio between the equilibrium concentrations of water in the plum pulp and in the plum peel
K_2	Equilibrium constant, defined by the ratio between the equilibrium concentrations of water in the plum pulp and in air vapour
k	Mass transfer coefficient defined by $k = \frac{D_2}{\delta K_1}$
L	Parameter defined by $L = \frac{R_1 D_2}{\delta K_1 D_1}$
δ	Peel thickness (m)

Subscripts

0	Refers to time $t = 0$
1	Refers to plum pulp
2	Refers to plum peel
3	Refers to air surrounding the plum
eq	Refers to equilibrium conditions ($t \to \infty$)
∞	Refers to air bulk

Superscripts

TR	Refers to treated plums: abraded (Abr) or dipped (EtOl)
UT	Refers to untreated plums
WP	Refers to plums with no peel

region, Italy. Drying experiments were carried out in a convection oven at 60 °C, with an air speed of 0.5 m/s, so as to reduce the average moisture of plums to about 0.25% w/w. Before drying, samples of about 20 plums (average weight 61.6 g), were submitted to one of the following pre-treatments (TR):

a. abrasion (Abr) in a pilot plant, constituted primarily of a cylinder containing abrasive material internally. The speed of rotation of the cylinder was variable and its energy was supplied by an electric motor. Different types of abrasive materials were tested to achieve a light abrasion, avoiding break-ups of the peel and loss of liquid during drying. During the preliminary study, it was determined that better operating conditions for the pre-treatment were obtained by adopting the following parameters: abrasion with paper type PW400; speed of cylinder' rotation 120 rpm; time for the abrasive treatment 15 min;
b. immersion in an aqueous solution of 2% (v/v) ethyl oleate and 2.5% (v/v) K_2CO_3 at 40 °C for 5 min (EtOl);
c. untreated samples as reference (UT).

3. Results and discussion

3.1. General

At the end of the drying process, carried out at 60 °C so as to reduce the plums' quality loss, the original structure of all prunes was maintained, independently of the pre-treatment used. The abrasion did not involve any loss of juice since not one crack was observed either after the physical pre-treatment or after drying. The proposed physical pre-treatment, without altering in a meaningful way the other qualitative characteristics of the plums, most importantly reduced the dehydration time considerably and, as a result, caused only a small loss of nutritive substances in the prunes. A more in-depth analysis of the prunes' qualitative changes, assessed by analysing the changes in skin colour, sugars by HPLC, total phenols, total anthocyanins, and reactive substances to the vanillin-HCl, constitutes the object of our second research paper (Part 2).

3.2. Mathematical model of plum dehydration

In the dehydration process of plums by means of warm air, simultaneous heat and water transport take place. Since the duration of the thermal transient was generally found to be far less than the duration of the dehydration process, mass transport may be regarded as taking place under isothermal conditions. In other words, the whole drying process is controlled by mass transport only, as for similar processes (Bird, Stewart, & Lightfoot, 1960; Di Matteo, Cinquanta, Galiero, & Crescitelli, 2000; Peri & Riva, 1984). In the section which follows the mathematical model will be first described in the hypothesis that the radius of plums is constant during the entire dehydration process, following which this hypothesis will no longer be employed.

3.3. Model for plum with constant radius

Under the assumptions that pulp and peel (if present) are uniform and isotropic, and the plums are spherical with constant radius, the mathematical model of plum dehydration can be reduced to that of mass diffusion from a spherical body surrounding an impermeable sphere (the plum-stone) (Bird et al., 1960; Carslaw & Jaeger, 1980; Crank, 1975; Luikov, 1968).

Fig. 1 shows the schematic water concentration profiles in the plum pulp and peel, as well as in the gaseous film surrounding the plum.

The mathematical model that describes the diffusion of water through whole plums must account for its diffusion, both in the pulp and in the peel. Both processes are described by the mathematical model:

$$\frac{\partial c_i}{\partial t} = D_i\left(\frac{\partial^2 c_i}{\partial r^2} + \frac{2}{r}\frac{\partial c_i}{\partial r}\right), \tag{1}$$

where the index $i = 1$refers to the pulp [i.e. for $r \in (R_0, R_1)$] and $i = 2$ to the peel [i.e. for $r \in (R_1, R_2)$], D_1 is the water diffusivity in the plum pulp, which is much higher than that in the plum peel (D_2).

The evolution of water concentration within plum pulp can be estimated by solving the differential Eq. (1) for $i = 1$with the following initial (IV) and boundary conditions (BV):

$$\begin{aligned} &\boldsymbol{IV}: c_1(r,0) = c_{10},\ \ r \in [R_0, R_1],\ \ t = 0, \\ &\quad \frac{\partial c_1}{\partial r} = 0,\ r = R_0,\ t > 0, \\ &\boldsymbol{BV}: D_1\frac{\partial c_1}{\partial r} = D_2\frac{\partial c_2}{\partial r},\ r = R_1,\ t > 0, \\ &\quad c_1(R_1,t) = K_1 c_2(R_1,t),\ t > 0, \end{aligned} \tag{2}$$

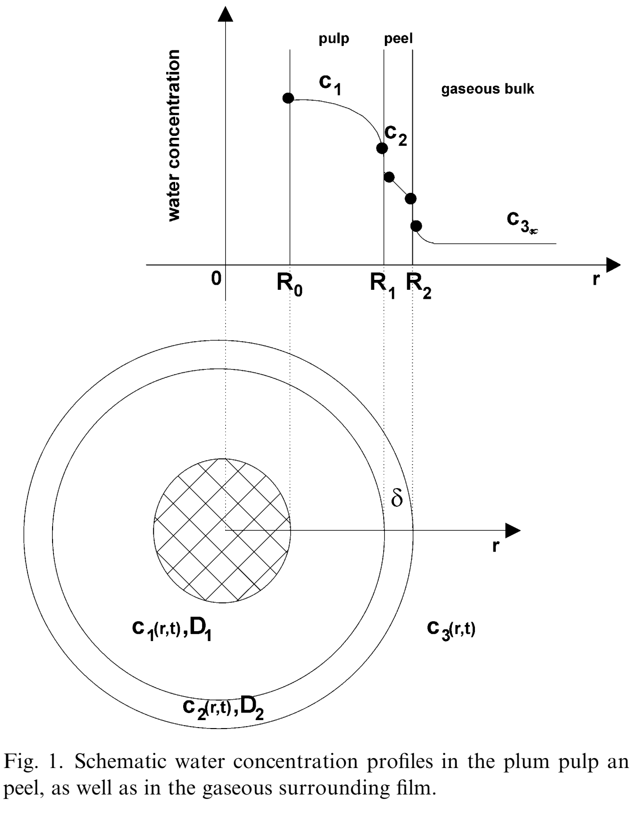

Fig. 1. Schematic water concentration profiles in the plum pulp and peel, as well as in the gaseous surrounding film.

In the above Eq. (2) the equilibrium–distribution curve relating water concentration in the pulp/peel interface was assumed to be a linear one and characterised by the equilibrium constant K_1.

Similarly, the evolution of water concentration within the plum peel can be obtained by solving Eq. (1) for $i = 2$ with the following initial and boundary conditions:

$$\begin{aligned} &\boldsymbol{IV}: c_2(r,0) = \frac{c_{10}}{K_1},\ \ r \in [R_1, R_2],\ \ t = 0, \\ &\boldsymbol{BV}: -D_2\frac{\partial c_2}{\partial r} = h\left[\frac{c_2(R_2,t)}{K_2} - c_{3\infty}\right],\ \ r = R_2,\ t > 0 \end{aligned} \tag{3}$$

In the above Eq. (3), the equilibrium–distribution curve relating water concentrations at peel/outer environment interface was assumed to be a linear one and characterised by the equilibrium constant K_2. Moreover, water diffusion in the gaseous film around the plum was described by means of the convective mass transfer coefficient h (Bird et al., 1960).

Owing to the thinness of plum peel, it is possible to neglect water accumulation in the peel, thus considering a linear steady-state distribution of $c_2(r,t)$in the peel for any time. Moreover, because in the experiments described here, the gaseous velocity was quite high, the resistance to the mass transport out of the plums can be neglected (i.e. $h \to +\infty$). In this case, the boundary condition for $r = R_2$ becomes:

$$c_2(R_2,t) = K_2 c_{3\infty}\quad t > 0,$$

and the mathematical model can be reduced to Eq. (1), with $i = 1$, with the following initial (IV) and boundary conditions (BV):

$$\boldsymbol{IV}: c_1(r,0) = c_{10}, r \in [R_0, R_1],\ \ t = 0,$$

$$\begin{aligned} &\boldsymbol{BV}: \frac{\partial c_1}{\partial r} = 0, r = R_0, t > 0, \\ &\quad -D_1\frac{\partial c_1}{\partial r}\Big|_{r=R_1} = k\big(c_1(R_1,t) - c_{1\mathrm{eq}}\big) \end{aligned} \tag{4}$$

where $k = \dfrac{D_2}{\delta K_1}$.

For peeled plums, the dehydration process can be mathematically described by accounting only for the diffusion of water in the plum pulp and steam in the gaseous film surrounding the plum berry. The diffusion of water in the pulp (if $h \to \infty$) is described by the model (1) with $i=1$ and the initial and boundary conditions become:

$$\boldsymbol{IV}: c_1(r,0) = c_{10},\ r \in [R_0, R_1],\ t = 0,$$

$$\boldsymbol{BV}: \frac{\partial c_1}{\partial r} = 0,\ r = R_0,\ t > 0,$$

$$c_1(R_1, t) = c_{1\text{eq}},\ r = R_1,\ t > 0.$$

The thermodynamic equilibrium at the interface gas/plum pulp is expressed as a linear law:

$$c_{1\text{eq}} = K_3 c_{3\infty}.$$

With these hypotheses, an analytical solution can be obtained by following the same methodologies as used by Carslaw and Jaeger (1980) and Crank (1975) for other geometries.

In another work, just submitted, the analytical solutions are derived for water concentration and humidity evolution. It is shown that the two solutions can be expressed in the same form:

$$\frac{H - H_{\text{eq}}}{H_0 - H_{\text{eq}}} = \frac{6}{R_1^3 + R_0^3} \sum_{n=1}^{\infty} (-MI)_n \exp\left\{-\frac{D_1 \beta_n^2 t}{R_1^2}\right\} \tag{5}$$

where

- for berries with peel

$$(-MI)_n = \frac{R_1\left[(L-1)^2+\beta_n^2\right]}{(1-R_0/R_1)\left(R_1^2+R_0^2\beta_n^2\right)\left[(L-1)^2+\beta_n^2\right]+\left[(L-1)R_0+R_1\right]\left[(L-1)R_1+R_0\beta_n^2\right]} \times \left[\frac{L\left[R_1^2+R_0^2\beta_n^2\right]}{\beta_n\left[(L-1)-\frac{R_0}{R_1}\beta_n^2\right]}\right]^2 \cos^2[\beta_n(1-R_0/R_1)]$$

- for berries without peel

$$(-MI)_n = \left(\frac{R_1^2+R_0^2\beta_n^2}{\beta_n}\right)^2 \frac{R_1^2}{R_1^3+(R_1-R_0)R_0^2\beta_n^2} \times \cos^2[\beta_n(1-R_0/R_1)]$$

For both of them, the following holds:

$$\beta_n = R_1 \alpha_n$$

$$L = \frac{R_1 h_2}{d_2} = \frac{R_1 D_2}{\delta K_1 D_1}$$

3.4. Model for plum with variable radius

In the dehydration process, the radius of the plum changes considerably. In fact, from the data shown in Table 1, it can be deduced that the radius changes by more than 100% during the process. For this reason, the hypothesis of constant radius during the dehydration process is not reliable and radius variation must be taken into account. This is accomplished by using the above equations to compute the evolution of the water concentration for a time interval Δt, small enough to consider the radius constant. To follow the entire dehydration process, the same equations are used at successive time intervals, each one with a different value for the radius:

- because the radius of the plum changes during the *i*-th time interval, Δt the average between the initial and final values, is considered as the radius to be used in the model;
- the humidity obtained at the end of the ($i-1$)th time interval is taken as the initial water content for the *i*-th time interval; and
- initial water concentration profile is computed by keeping constant the water content obtained at the previous step.

3.5. Parameter estimation

The values of unknown parameters of the models described here were estimated by fitting the experimental drying data collected when using peeled, untreated and treated plums. The unknown parameters

Table 1
Physical changes of different pretreated plums during dehydration

Drying time (h)	d[a] TQ	l[b] TQ	d Abr	l Abr	d EtOl	l EtOl
0	46.5	48.0	45.8	48.8	49.0	50.0
4	44.3	46.5	42.8	43.3	47.2	47.2
6	43.4	46.0	40.6	41.4	45.8	45.8
8	42.5	45.8	38.9	40.0	44.4	43.8
10	41.7	45.7	36.6	38.5	41.9	41.9
12	40.8	45.0	34.9	37.0	41.0	41.0
14	39.7	44.5	32.4	34.6	39.1	39.7
16	39.3	44.4	29.9	32.0	38.0	37.5
18	38.7	43.2	27.3	30.8	36.6	34.4
20	38.3	42.2	24.6	28.8	35.3	33.4
22	37.5	42.0	23.2	27.2	34.1	31.1
24	37.1	41.5	22.1	26.6	32.3	27.7
26	36.1	40.8	21.1	25.4	29.5	27.1
28	34.7	40.0	20.1	24.6	28.0	25.8
30	33.0	38.8	18.8	23.0	25.0	22.6
32	30.7	37.7	18.4	22.0	23.8	21.3
34	29.0	37.0	18.0	21.5	22.8	20.0
36	28.2	36.0	17.7	20.7	22.0	19.7

[a] d, diameter of plums ($m*10^{-3}$).
[b] l, length of plums ($m*10^{-3}$).

to be estimated are the following: the water diffusivity in the pulp (D_1), the water equilibrium concentration ($c_{1\text{eq}}$) and the mass transfer coefficient in the plum peel (k) for the untreated or treated samples.

A first assessment of such parameters was performed as follows:

1. The equilibrium values of the moisture content ($H_{1\text{eq}}$) for all plum berries used were estimated by averaging the experimental humidity determined at the end of each drying process on the assumption that such mean values coincided with the equilibrium ones ($H_{\text{eq}}^{\text{WP}}$,$H_{\text{eq}}^{\text{UT}}$,$H_{\text{eq}}^{\text{TR}}$). From these values, $c_{1\text{eq}}$ for plums without peel ($c_{1\text{eq}}^{\text{WP}}$), for untreated plums ($c_{1\text{eq}}^{\text{UT}}$), and for treated plums ($c_{1\text{eq}}^{\text{TR}}$), can be easily estimated by considering experimental plum volumes.
2. The diffusivity of water in the plum pulp (D_1) can be easily estimated by considering just the first term in the series (5) and by describing the logarithmic reduction of its first term as a linear function of time

$$\ln\left(\frac{H - H_{\text{eq}}}{H_0 - H_{\text{eq}}}\right) = \ln\left(\frac{6}{R_1^3 - R_0^3}\right) + \ln\left((-MI)_1\right) - \frac{D_1 \beta_1^2 t}{R_1^2} \tag{6}$$

   and using the earlier $H_{\text{eq}}^{\text{WP}}$ end value and the experimental $H - t$ data pertaining to peeled plum berries. Such a parameter was also used to describe water diffusivity in the plum pulp of all the samples tested, the latter always being composed of plum berries of the same variety, that were almost simultaneously harvested in the same region. It must be pointed out that Eq. (6) should be used only for large values of t, ($t \rightarrow \infty$): in fact, for $t = 0$ it does not forecast $H \rightarrow H_0$.
3. The starting value of the mass transfer coefficient, k, which accounts for water diffusivity in the untreated or treated plums (D_2) and equilibrium constant at pulp/peel interface (K_1), was obtained by plotting the corresponding humidity ratio $\left(H - H_{\text{eq}}\right)/\left(H_0 - H_{\text{eq}}\right)$ vs. time, by numerically estimating its slope for $t = 0$ and by equating such a slope to the derivative of the above humidity ratio with respect to time as calculated from Eq. (5) and computed by accounting only for the first term of the series and for $t = 0$:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{H - H_{\text{eq}}}{H_0 - H_{\text{eq}}}\right) = -\frac{2}{R_1^2 \frac{\mathrm{d}R_1}{\mathrm{d}t} - R_0^2 \frac{\mathrm{d}R_0}{\mathrm{d}t}} \frac{\mathrm{d}(-MI)_1}{\mathrm{d}t} \frac{D_1 \beta_1^2}{R_1^2}$$

Since the parameter k is used to define L and thus, implicitly, β_1, its estimate has to be obtained by means of successive iterations.

The optimal estimates of all unknown parameters $\left(D_1, k^{\text{UT}}, k^{\text{TR}}, c_{1\text{eq}}^{\text{WP}}, c_{1\text{eq}}^{\text{UT}}, c_{1\text{eq}}^{\text{TR}}\right)$, were derived by minimising the following performance index:

$$\Phi = \sum_{i=1}^{N} \left(H_i^S - H_i^T\right)^2$$

which represents the sum of the squared differences among the experimental humidity H_i^S data referred to peeled, untreated and treated plum berries and those calculated via their corresponding H_i^T models. Moreover, any water content was forecast by considering the first n terms of any series only, n being sufficiently high to assure a relative error of less than 1% between the generic n and $(n + 1)$ terms summation. In all the forecasts computed, a value of $n = 4 \div 5$ was sufficient.

The minimisation exercise was carried out by using a non-linear estimation method based on a mixed (direct/gradient) algorithm (Buzzi Ferraris, 1972), thus leading to the optimal values of the earlier unknown parameters. Their covariance matrix was also computed to determine the confidence intervals of all the estimates at a 95% confidence level (Bard, 1974; Buzzi Ferraris, 1972).

The continuous lines in Fig. 2 show the calculated time variation in moisture content for plums tested and show quite a good agreement with the corresponding experimental values.

The optimal value of the water diffusion coefficient in the pulp is equal to

$$D_1 = 2.648 \times 10^{-6} \pm 1 \times 10^{-9} \ \left(\text{m}^2/\text{h}\right)$$

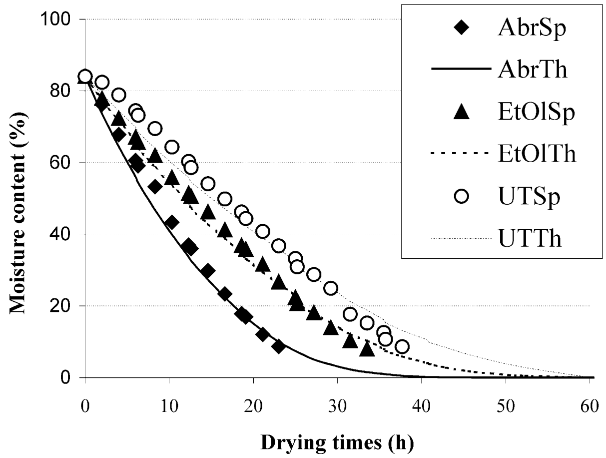

Fig. 2. Experimental (Sp), and theoretical (Th) values of moisture changes (%) vs. drying times for plums, the peel of which was untreated (UT) or pre-treated by dipping into ethyl oleate (EtOl) or by abrasion (Abr).

Optimal values of transfer coefficients are:

$$k^{\mathrm{TR}} = 3.68961 \times 10^{-4} \pm 0.01 \times 10^{-4}\ (\mathrm{m/h})\ \text{chemically treated}$$

$$k^{\mathrm{TR}} = 6.78933 \times 10^{-4} \pm 0.006 \times 10^{-4}\ (\mathrm{m/h})\ \text{mechanically treated}$$

$$k^{\mathrm{TR}} = 2.50524 \times 10^{-4} \pm 0.002 \times 10^{-4}\ (\mathrm{m/h})$$

The mass transfer coefficients in the peel plum treated k^{TR}, both chemically and mechanically, were obviously found to be greater than the untreated one $k^{\mathrm{UT}}$and measure the greater capability of the pre-treatments used to enhance water diffusivity in the plum peel relative to that in the untreated samples. The greater efficacy of the mechanical treatment than the chemical one is noteworthy.

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